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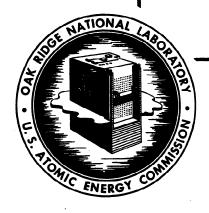
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CRITICAL MASS STUDIES

PART VIII

AQUEOUS SOLUTIONS OF U²³³

J. K. Fox L. W. Gilley E. R. Rohrer



OAK RIDGE NATIONAL LABORATORY

operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

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CRITICAL MASS STUDIES, PART VIII AQUEOUS SOLUTIONS OF U²³³

J. K. Fox, L. W. Gilley and E. R. Rohrer

SEP 23 1959

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
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ABSTRACT

A series of experiments have been performed to establish the critical parameters of aqueous solutions of uranyl nitrate and uranyl fluoride in which the uranium contained 98.7% U233. Solutions were made critical in both spherical and cylindrical geometries with paraffin or water as a neutron reflector and, in two instances, with no reflector. The U²³³ concentration varied from 30 to 600 g/liter. The minimum critical mass observed was 590 g of U^{233} in the solution having an H:U233 atomic ratio of 419 occupying a 10.4-in.-dia sphere. The minimum measured volume was 3.66 liters in a 6.7-in. equilateral cylinder containing a solution with an $H:U^{233}$ atomic ratio of 39.4. Extrapolated source neutron multiplication data indicate that a 5-in.-dia cylinder can be made critical if reflected, but a 4-in.-dia cylinder would be subcritical at all moderations. It was also found that 2.02 kg of U^233 in an unreflected 10-in. equilateral cylinder is critical with a solution having an $H:U^{233}$ atomic ratio of 154. An unreflected sphere 12.6 in. in diameter is critical with 1.14 kg of U²33 in a solution with an H:U²33 ratio of 381. Extension of the data to geometries other than those used experimentally was made by an empirical calculation.

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INTRODUCTION

Nuclear safety in processing plants for fissionable materials can be imposed by limiting equipment dimensions, batch sizes, and other plant variables in accordance with the results of experimental investigations of the critical parameters of the fissionable material of interest. Experiments of this kind have been performed at Hanford with plutonium and have been in progress at Oak Ridge with U235 for some time, but the unavailability of sufficient amounts of U233 has prohibited any study of this isotope. A limited experimental program became possible, however, when approximately 2.5 kg of isotopically pure U233 was released to the ORNL Critical Experiments Facility in 1952. A series of critical experiments were performed in which the U233 was used in aqueous solutions contained in cylindrical and spherical vessels and reflected with paraffin or water. The solution concentrations varied from 30 to 600 g of U233 per liter of solution. It was not possible to make all of the attempted experiments critical because of the limited inventory of U233 or the limited capacity of some of the test vessels. Source neutron multiplication measurements in these subcritical experiments were extrapolated to give estimates of the critical conditions. It was also possible to make two unreflected vessels critical.

A summary of all of these measurements and an extension of them by an empirical calculation has provided some design basis for equipment for processing the $U^{2\bar{3}\bar{3}}$ isotope.

I. EXPERIMENTAL APPARATUS AND PROCEDURE

The uranium used in these experiments contained $98.7\%~U^{233}$, the remainder being about equal quantities of U^{234} and U^{238} . It was first used as an aqueous solution of uranyl nitrate $(UO_2(NO_3)_2)$ and later as a solution of the more soluble uranyl fluoride (UO_2F_2) in order to extend the measurements to higher chemical concentrations and to reduce the neutron absorption. The solutions were contained in cylinders having diameters between 4-1/2 and 12 in. and in spheres having diameters of 10.4 and 12.6 in. The vessels in most of the experiments had effectively infinite hydrogenous reflectors, but in two cases it was possible to achieve criticality with no reflector. The critical assemblies were of simple geometry and relatively free of extraneous neutron absorbers.

^{1.} F. E. Kruesi, J. O. Erkman and D. D. Lanning, "Critical Mass Studies of Plutonium Solutions," HW-24512 (1952).

^{2.} See, for example, J. K. Fox, L. W. Gilley and D. Callihan, "Critical Mass Studies, Part IX, Aqueous U²³⁵ Solutions," ORNL-2367 (1958).

Since neutron moderation was one of the principal variables investigated, the chemical concentrations of the solutions were altered in the course of the experiments and $\text{H}:\mathbb{U}^2$ atomic ratios were determined from uranium analyses. The uranyl nitrate solution was shown to contain some excess nitric acid, equivalent to a total $\text{H}:\mathbb{U}^2$ atomic ratio of 2.66. Some of the properties of the solutions, including the results of spectrochemical analyses for metallic impurities resulting from corrosion of the equipment, are given in Appendix A.

The solution vessels were constructed of type 3S aluminum and coated with thin layers of either Heresite or Unichrome to reduce corrosion. Neither the Heresite nor the Unichrome contains any significant neutron-absorbing metallic impurities, although Unichrome is about 30 wt% chlorine and is thus a weak neutron absorber. Estimates of the effect of the Unichrome on the experimental results are noted in the recorded data.

The cylinders having diameters less than 6.7 in. were 36 in. high, while those with diameters of 6.7 to 12 in. were approximately equilateral. The cylinders of larger diameter and the spheres were mounted in an outer cylinder of sufficient capacity to provide an effectively infinite water or paraffin neutron reflector completely surrounding the vessel under study. The smaller diameter cylinders had reflectors on the sides and bottom but not on the top. Neoprene gaskets were used to seal the cylinder covers.

Immediately adjacent to the periphery of each cylinder was a water-filled annular section into which a cadmium sheet, normally suspended from a magnet, could be inserted as a safety operation. A photograph of one of the vessels without a reflector is shown in Fig. 1. A neutron source, located in the reflector region, was positioned by remote control.

The solution flowed into the cylinders or vessels from a storage manifold which consisted of five 3-in.-dia type 347 stainless steel pipes mounted vertically on a bracket and spaced on 15-in. centers with their axes coplanar as shown in Fig. 2. The five pipes were each 37 in. long and were connected by smaller pipes at both the top and the bottom. The manifold had a total capacity of 23 liters and was connected by flexible tubing and diaphragm-type valves at the top and bottom of the vessel under test, thereby forming a closed transfer system. This design, which is shown in schematic form in Fig. 3, reduced health hazards and utilized the limited inventory of U²³³ as effectively as possible. The manifold was suspended from a rack which could be moved vertically by a remotely controlled motor. The ability to lower the manifold to remove solution from a test vessel served as an additional safety device.

The radiation detection instrumentation was conventional and included four parallel channels arranged to operate the safety devices. The detectors in three of these channels were BF_3 -filled ionization chambers with signal amplification by dc amplifiers or vibrating reed electrometers. The signals were observed on strip chart recorders. The fourth circuit consisted of an anthracene crystal and a photomultiplier tube. While approaching critical, the apparent neutron source multiplication was measured by boron-lined proportional counters.

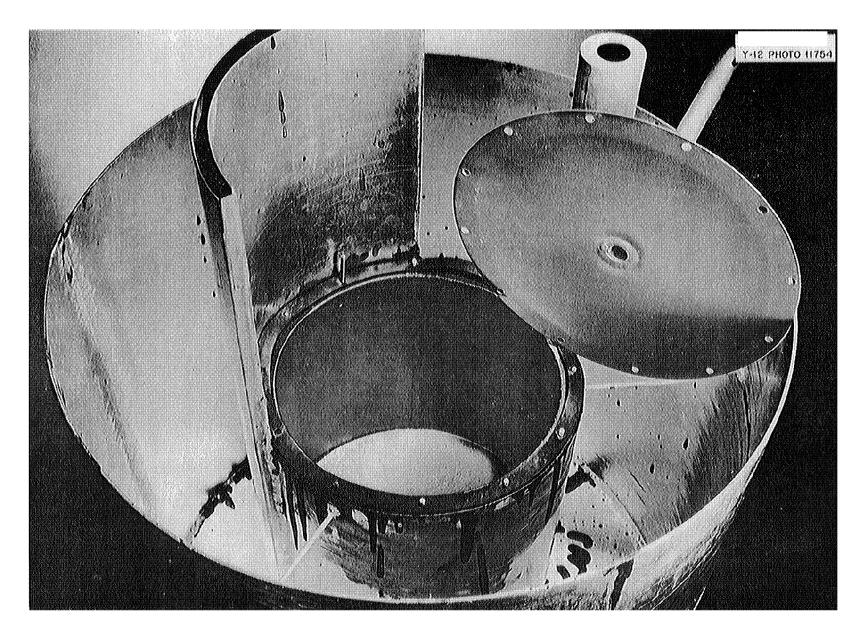


Fig. 1. Typical Containing Vessel for Experiments with Aqueous Solutions of U^{233} . U^{233} solution was contained in the inner cylinder and surrounded by a paraffin or water reflector.

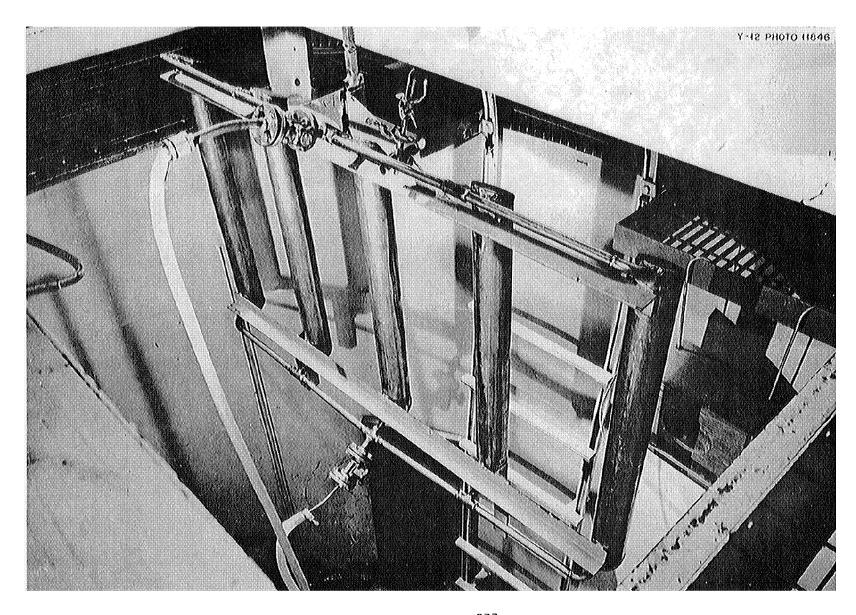


Fig. 2. Storage Manifold for Aqueous Solutions of U^{233} . The five pipes are 3 in. in diameter, 37 in. long, and mounted 15 in. apart.

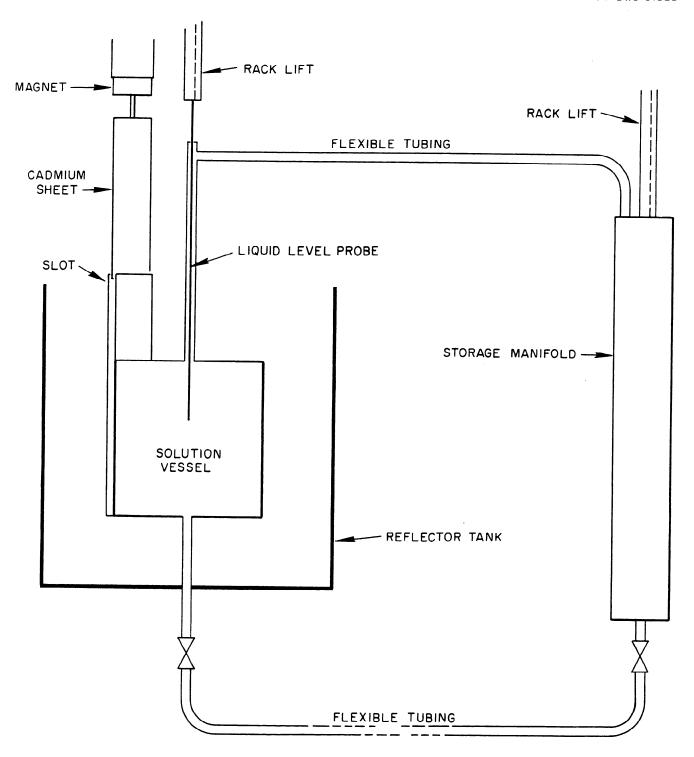


Fig. 3. Arrangement of Solution Vessel and Storage Manifold for Critical Experiments with Aqueous Solutions of U $^{233}\,$

In the uranyl nitrate solution experiments, the liquid level in a vessel was determined from an empirical calibration curve relating the vertical position of the manifold to the areas of the manifold and the solution vessel. In the uranyl fluoride solution experiments the position of the liquid level in a vessel was indicated by a selsyn connected to a motor-driven electrical contact inserted through a rubber diaphram across the tube connected to the top of the vessel.

The procedure in these experiments was similar to that used previously 2 with solutions of U^{235} , although the smaller delayed neutron yield of U^{235} required a more careful approach to critical. An effort was made in each case to adjust the concentration in order to have the system critical when full.

II. EXPERIMENTAL RESULTS

The critical data obtained in these experiments are tabulated in Appendix B along with estimates of the critical parameters derived from the source neutron multiplication curves of those assemblies which could not be made critical because of limitations imposed by the available quantity of $U^{2\bar{3}\bar{3}}$ or by the capacities of the containers. The results from critical assemblies and from the extrapolations in which there is greatest confidence are summarized in Figs. 4, 5, and 6.

Figure 4 is a plot of the critical mass as a function of the critical volume of reflected vessels. The data for uranyl nitrate and uranyl fluoride solutions contained in approximately equilateral cylinders can be represented by the same curve for the more dilute solutions, but, in the more concentrated range, the effect of the nitrate ion on the critical dimensions is measurable. This difference in critical masses at higher concentrations may be due to the lower hydrogen density in the uranyl nitrate solution. The U^{233} density in the two solutions is shown as a function of the $H:U^{233}$ ratio in Fig. A-l in Appendix A.

The critical volumes and critical masses measured in spheres and essentially equilateral cylinders, each surrounded by a neutron reflector, are plotted in Figs. 5 and 6, respectively, as functions of the concentration of the solutions expressed as the $\text{H}: \text{U}^2$ 33 atomic ratio. The curves in each figure have been drawn through the points describing the cylinders.

The two experiments without neutron reflectors used a 10-in.-dia equilateral cylinder and a 12.6-in.-dia sphere. The critical data for these two unreflected vessels are included in Appendix B.

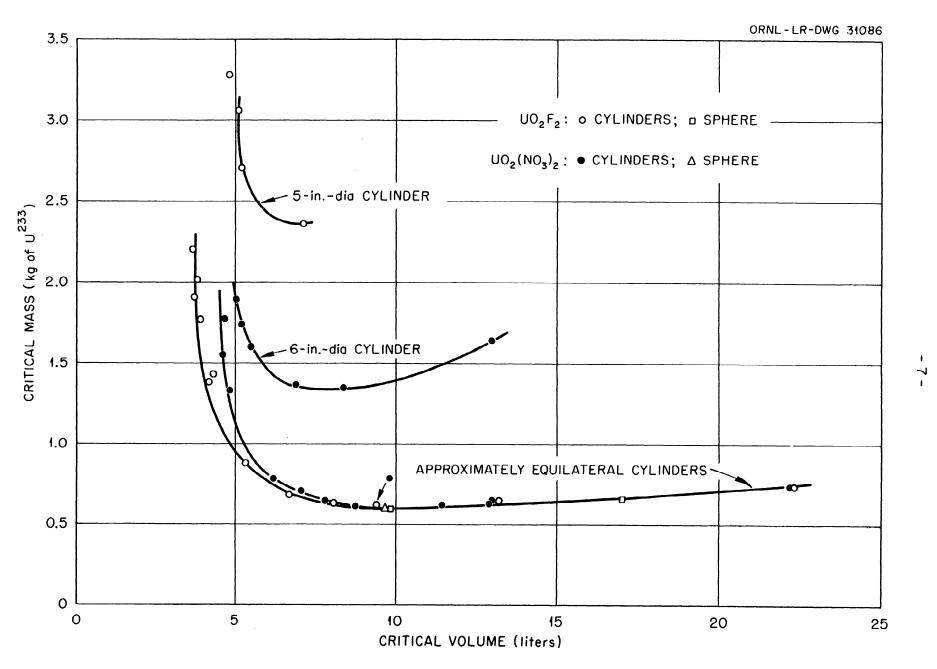


Fig. 4. Critical Mass as a Function of Critical Volume of Reflected Vessels Containing Aqueous Solutions of U^{233} .

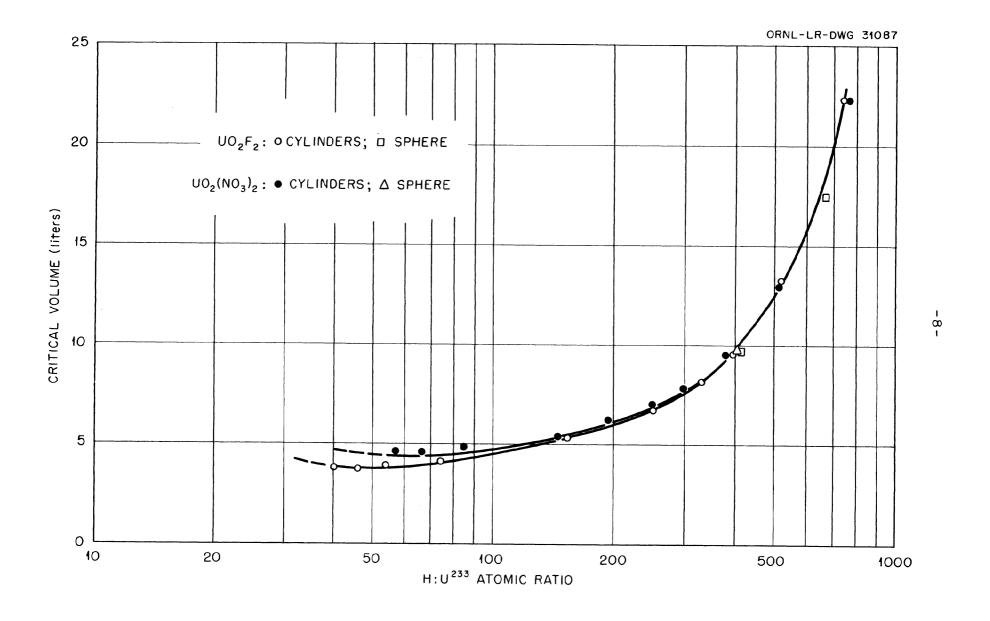


Fig. 5. Critical Volume as a Function of the H:U²³³ Atomic Ratios of Solutions Contained in Reflected Equilateral Cylinders and Spheres

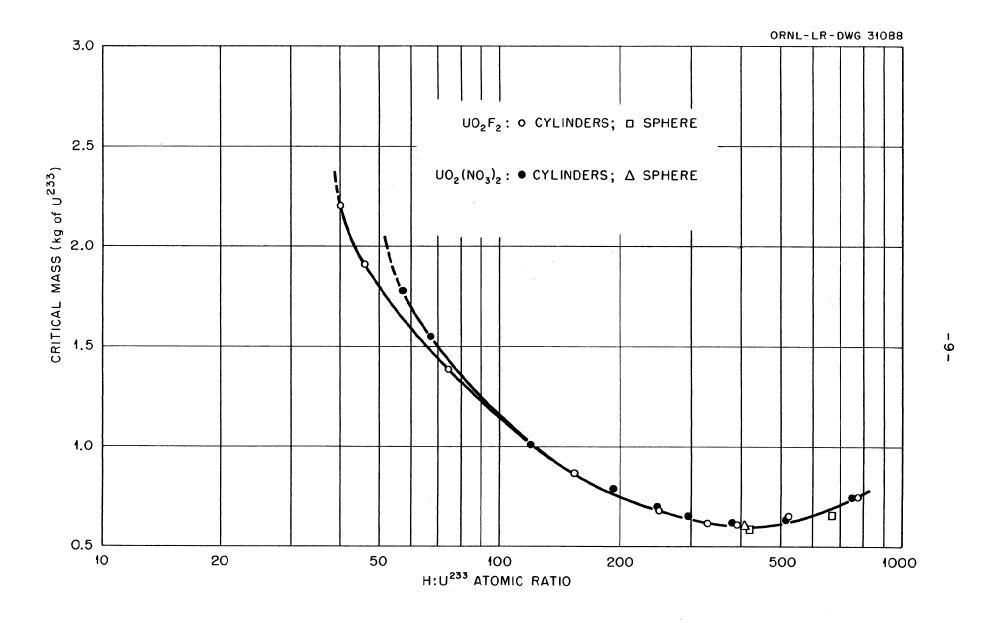


Fig. 6. Critical Mass as a Function of the H:U²³³Atomic Ratios of Solutions Contained in Reflected Equilateral Cylinders and Spheres

III. ACCURACY OF THE MEASUREMENTS

The results of these experiments are subject to the usual errors in uranium analyses and solution densities and in the calibration of the vessel capacities, all estimated to be ±0.5%. There are, however, two additional sources of error which broaden the uncertainty. In the experiments in which uranyl nitrate solutions were used the indirect method of measuring the height, and hence the volume, of the solution in the vessels makes the results less certain than those from later experiments with uranyl fluoride in which the position of the liquid surface was measured directly.

An error in the experiments with the uranyl fluoride solutions is a bias resulting from the properties of a corrosion-inhibiting coating material with which one spherical and three cylindrical vessels were lined. This coating was a polyvinyl chloride plastic, Unichrome, which is about 30 wt% chlorine. It is difficult to evaluate the increase in critical mass due to the absorption of neutrons by the chlorine because the effect is a function of geometry. It is noted in Table B-1, however, that the critical volume at a particular concentration (H:U233 = 74.1) in a 14-cm-dia cylinder lined with Unichrome is about the same as that in a 12.7-cm-dia cylinder lined with a phenol base plastic, Heresite, whereas it would be expected to be somewhat less. After the discovery of the chlorine impurity, one definitive experiment was performed to determine the effect of a Unichrome liner on the critical concentration of a U235 uranyl fluoride solution in a sphere 32 cm in diameter. A 2% decrease in the concentration was observed upon removal of the Unichrome.

In summarizing the question of accuracy, it is believed that the results from critical experiments with uranyl fluoride in Heresite-lined vessels are good to $\pm 1\%$; data from the same vessels with uranyl nitrate have an uncertainty of no more than $\pm 3\%$. The critical mass measured in the 32-cm-dia sphere is high by about 2% because of a bias. The masses in the 14- and 17-cm-dia cylinders are too high by an amount not well known but probably less than 10%.

Extrapolations have been made of the source neutron multiplication curves obtained in those tests which could not be made critical because of inventory or geometric limitations. The critical parameters obtained in this manner are given in the tabulated results with the maxima from which the extrapolations were made. The accuracy of these values is strongly dependent upon the length of the extrapolation.

Appendix A

COMPOSITION OF URANIUM SOLUTIONS

Table A-1. U²³³ Concentrations in Solutions

Solution	_Մ 233 Co	ncentration
Density (g/cc)	g per g of Solution	g per cc of Solution
In UO2(NO3)2 Soluti	ion with $N:U^{233} = 2.66$	
1.697	0.289	0.490
		0.381
1.480	•	0.336
	•	0.275
1.287		0.198
		0.167
		0.160
		0.127
		0.117
1 145		0.101
		0.084
		0.070
		0.067
		0.063
		0.062
		0.055
		0.049
		0.044
		0.040
1.046	0.032	0.033
In UO ₂ F ₂ Solution		
1.801	0.380	0.684
	0.352	0.600
1.625	0.327	0.531
1.604		0.519
1.592		0.503
1.530	0.295	0.451
1.388	0.239	0.332
1.199	0.138	0.165
1.198	0.138	0.165
	0.091	0.102
1.090	0.071	0.078
1.079	0.062	0.067
	c.061	0.066
	0.060	0.065
		0.061
		0.060
	0.047	0.049
	· · · · · · · · · · · · · · · · · · ·	0.039
1.035	0.032	0.033
	Density (g/cc) In UO ₂ (NO ₃) ₂ Solution 1.697 1.543 1.480 1.394 1.287 1.238 1.232 1.185 1.165 1.145 1.101 1.093 1.090 1.087 1.077 1.069 1.061 1.056 1.046 In UO ₂ F ₂ Solution 1.801 1.707 1.625 1.604 1.592 1.530 1.388 1.199 1.198 1.121 1.090 1.076 1.075 1.076 1.075 1.071 1.070 1.059 1.043	Density (g/cc) g per g of Solution In UO ₂ (NO ₃) ₂ Solution with N:U ²³³ = 2.66 1.697 0.289 1.543 0.247 1.480 0.227 1.394 0.197 1.287 0.154 1.238 0.135 1.232 0.130 1.185 0.107 1.165 0.100 1.145 0.088 1.121 0.075 1.101 0.064 1.093 0.061 1.090 0.058 1.087 0.057 1.077 0.051 1.069 0.046 1.061 0.041 1.056 0.032 In UO ₂ F ₂ Solution 1.801 0.380 1.707 0.352 1.625 0.327 1.604 0.324 1.592 0.316 1.592 0.316 1.530 0.295 1.388 0.239 1.199 0.138 1.198 0.138 1.198 0.138 1.198 0.138 1.199 0.062 1.076 0.061 1.079 0.062 1.076 0.061 1.075 0.060 1.071 0.057 1.070 0.056 1.059 0.047 1.043 0.037

Table A-2. Isotopic Composition of Uranium in Solutions

	wt%	
Isotope	In UO ₂ (NO ₃) ₂ Solution	In UO ₂ F ₂ Solution
υ ² 33	98.7	98.7
_Մ 233 Մ ²³⁴	0.50	0.54
Մ ² 35 Մ ² 38	0.01	0.04
_U 238	0 . 79	0.72

Table A-3. Principal Impurities* in Solutions

	In $UO_2(NO_3)_2$	Solution	In UO2F2 Solution		
Element	Before Use	After Use	Before Use	After Use	
Al Ca	15 300	50 1500	400	8000	
Cr	10	15	8	250	
Fe	20	300	40	2500	
Mg	10	125	0	55	
Мо			0	200	
Na			650	100	
Ni	20	70	0	250	
S n			0	225	
Th	150				

^{*} Determined by spectrographic analysis.

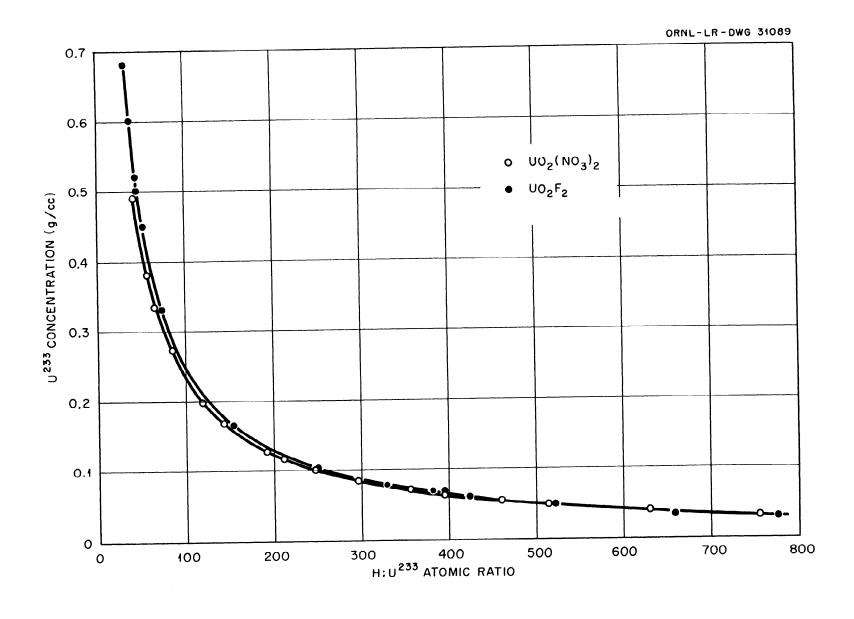


Fig. A-1. U^{233} Concentration in Aqueous Solutions of Uranyl Nitrate and Uranyl Fluoride as a Function of the $H:U^{233}$ Atomic Ratio.

Appendix B

TABULATIONS OF CRITICAL DATA

Table B-1. Critical Parameters of Uranyl Nitrate Solutions

(Numbers in parentheses represent extrapolated values derived from source neutron multiplication curves of subcritical assemblies.)

H:U ² 33		Critical Parameters		Maximum Values of Subcritical Assemblies			
Atomic Ratio ²	Reflector	Height (cm)	Volume (liters)	Mass (kg of U ² 33)	Height (cm)	Volume (liters)	Mass (kg of U ²³³)
			Cylinder :	12.7 cm (5.0 in.) in Dia	ameter ^b		
57.5 67.0 84.4 145	Paraffin " "	c c c		<u>.</u>	51 59 61 55	6.40 7.40 7.65 6.90	2.44 2.49 2.10 1.15
			Cylinder	15.1 cm (6.0 in.) in Di			
57.5 67.0 84.4 120 151 193	Paraffin	27.9 29.0 30.7 (38.5 ± 0.5 (46.8 ± 0.5 (73 ± 2	5.00 5.20 5.50 5) (6.9 ± 0.1) 5) (8.4 ± 0.1)	1.91 1.75 1.51 (1.37 ± 0.02) (1.34 ± 0.02) (1.65 ± 0.08)	36.8 45.4 55.4	6.60 8.15 9.80	1.30 1.31 1.24
			Cylinder 1	9.1 cm (7.5 in.) in Dia	meter		
57.5 67.0 145	Paraffin " "	16.3 16.2 18.6	4.65 4.60 5.30	1.77 1.55 0.89			
			Cylinder 2	0.5 cm (8.0 in.) in Dia	meter		
42.2 57.5 84.4 120 145 151 193 213 247	Paraffin " " " " " " "	(16.1 ± 0.2 14.4 14.7 16.4 16.7 16.7 18.8 19.3 (21.2 ± 0.2	4.75 4.85 5.40 5.51 5.51 6.20 6.37	(2.6 ± 0.03) 1.81 1.33 1.07 0.92 0.88 0.79 0.75 (0.70 ± 0.01)	14.0	4.60 6.56	2.25
		(81.8 = 0.		1.5 cm (8.5 in.) in Dian			
247 297	Paraffin	19.4 21.5	7.00 7.78	0.70 0.65			
			Cylinder 22	2.9 cm (9.0 in.) in Dia	meter		
356 379	Paraffin "	21.3	8.75 9. 3 9	0.62 0.62			
			Cylinder 25	5.5 cm (10.0 in.) in Di	ameter		
394 461 514 514	Paraffin "" Water	19.3 22.5 (25.2 ± 0. (25.5 ± 0.	9.82 11.45 1) (12.9 ± 0.05) 1) (13.0 ± 0.05)	0.63 0.63) (0.63 ± 0.03)) (0.64 ± 0.03)	25.1 25.1	12.77	0.63 0.63
			Cylinder 30	.5 cm (12.0 in.) in Dia	meter		
582 630 757	Paraffin "	21.1 23.8 30.4	15.40 17.40 22.20	0.68 0.70 0.75			
			Sphere 26.6	cm (10.4 in.) in Diame	ter		
405	Water	Full	9.66	0.60			

a. The compositions of the solutions are given in Appendix A.

b. No reflector on the top surface.

c. Apparently this assembly cannot be made critical at any height.

Table B-2. Critical Parameters of Uranyl Fluoride Solutions

(Numbers in parentheses represent extrapolated values derived from source neutron multiplication curves for subcritical assemblies.)

H:U ² 33		Critical Parameters		Maximum Values of Subcritical Assemblies			
Atomic Ratioa	Reflector	Height (cm)	Volume (liters)	Mass (kg of U ²³³)	Height (cm)	Volume (liters)	Mass (kg of U ²³³)
			Cylinder 11.	2 cm (4.5 in.) in Diame	ter ^{b,c}		
34.2	Paraffin	đ			29.9	2.95	2.02
39.4	**	đ			34.9	3.43	2.07
45.9	"	đ			42.6	4.19	2.18
53.7	11	ď			49.0 68.5	4.82 6.76	2.18 2.24
74.1		d	0.31.3	7 (5.0 in) in Diama		0.70	2.24
				7 cm (5.0 in.) in Diame			
34.2	Paraffin	(38 ± 2)	(4.8 ± 0.25		23.8 27.6	2.99 3.46	2.05 2.08
39.4	11	2.) (5.1 ± 0.25) (5.2 ± 0.1		32.4	4.07	2.11
45.9 74.1	"	$(56.5 \pm 0.)$) (5.2 ± 0.1 5) (7.1 ± 0.06		53.3	6.70	2.22
14.1		()0.) = 0.					
		(1.0 =		.7 cm (5.4 in.) in Diam		/ 53	0.05
74.1	Paraffin	(48.7 ± 0.		6) (2.37 ± 0.02)	46.3	6.77	2.25
				.1 cm (6.0 in.) in Diam	eter ^{D,C}		
74.1	Paraffin	24.0	4.31	1.43			
				.7 cm (6.6 in.) in Diam	eter ^C		
34.2	Paraffin	(20 ± 1			13.5	2.94	2.01
39.4	11	$(16.7 \pm 0.)$			16.3	3-55	2.13
45.2	11	$(17.4 \pm 0.$			16.9	3.69	1.96
45.9	"	16.9	3.67 2) (3.85 ± 0.0	1.91 4) (1.94 ± 0.03)	16.9	3.69	1.86
47.9	11	$(17.7 \pm 0.)$ $(18.0 \pm 0.)$			16.9	3.69	1.66
53.7 74.1	"	$(19.1 \pm 0.)$			16.9	3.69	1.23
			Cylinder 19	.1 cm (7.5 in.) in Diam	eter		
154	Paraffin	18.4	5.25	0.87			
			Cylinder 20	.5 cm (8.0 in.) in Diam	eter		
250	Paraffin	(20.2 ± 0.	05) (6.66 ± 0.0	2) (0.68 ± 0.02)	20.1	6:63	0.68
				.5 cm (8.5 in.) in Diam	eter		
329	Paraffin	(22.2 ± 0.	1) (8.04 ± 0.0	4) (0.65 ± 0.05)	21.6	7.80	0.61
			Cylinder 22	.9 cm (9.0 in.) in Diam	eter		
396	Paraffin	(23.1 ± 0.	1) (9.47 ± 0.0	4) (0.61 ± 0.05)	22.6	9.26	0.60
			Cylinder 25	.5 cm (10.0 in.) in Dia	meter		
522	Water	(25.9 ± 0.	1) (13.18 ± 0.0	5) (0.65 ± 0.05)	25.6	13.05	0.64
154	None		05)(12.22 ± 0.0	3) (2.02 ± 0.05)	23.8	12.15	2.01
775	Paraffin	30.5	Cylinder 30 22.28	0.5 cm (12.0 in.) in Dia	meter		
				6 cm (10.4 in.) in Diam	eter		
426	Water		(9.80 ± 0.1		Full	9.66	0.58
419	11	e	9.62	0.59			
390	11	f	9.28	0.61			
				9 cm (12.6 in.) in Diam	eter		
663	Water	Full	17.02	0.66			
<u> 381</u>	None	e	16.98	1.14			

a. The compositions of the solutions are given in Appendix A.

<sup>b. No reflector on the top surface.
c. Vessels coated with Unichrome; masses about 2% high because of impurities.</sup>

d. Apparently this assembly cannot be made critical at any height with the absence of a top reflector and the presence of Unichrome.

e. There was a 40-cm³ void above the critical solution.

f. There was a 380-cm³ void above the critical solution.

ESTIMATION OF CRITICAL PARAMETERS FOR UNREFLECTED SYSTEMS

The results from experiments performed almost exclusively with equilateral cylinders have been extrapolated to other cylindrical geometries using the method developed and applied by Bell³ to the extensive data on the critical parameters of U²³⁵ solutions. It is assumed that the age-theory critical equation can be made to fit experimental data from reflected and unreflected critical assemblies by proper choice of the extrapolation distances. In this analysis the reactivity is given by:

$$k_{eff} = \frac{\eta f e^{-\gamma B^2}}{(1 + L_{th}^2 B^2)} = \frac{\eta}{1 + \frac{\sigma_B(H)}{\sigma_B(U^{233})} C} \frac{e^{2\gamma B^2}}{(1 + L_{th}^2 B^2)}$$

where

$$L_{th}^{2} = \frac{L_{o}}{1 + \frac{\sigma_{\mathbf{a}}(U^{2}55)}{\sigma_{\mathbf{a}}(\mathbf{H})C}},$$

 L_0 = diffusion length of thermal neutrons in water, $C = H: U^{233}$ atomic ratio.

The constants used for the $U^{2\bar{3}\bar{3}}$ solutions are as follows:

$$L_0^2 = 8.2 \text{ cm}^2$$
,
 $\frac{\sigma_a(H)}{\sigma_a(U^{255})} = 0.328/575$,
 $\gamma = 27 \text{ cm}^2 \text{ (except as noted below)}$,
 $\eta = 2.33$

Values for the extrapolation distances for reflected and unreflected cylinders were derived from a comparison between calculated and experimental values of critical volumes of equilateral cylinders. These values are 8.25 cm and 4.95 cm for reflected and unreflected systems, respectively. The axial and radial extrapolation distances were assumed equal in calculating the buckling. It was found necessary to apply a multiplying factor (1.16) to the ratio of the thermal absorption cross sections of hydrogen and U²³³ in order to have both % and the extrapolation distance as constants and still fit the uranyl fluoride data in the high concentration region. It was also necessary to arbitrarily vary the neutron age with concentration, as shown in Fig. C-1, to obtain agreement with the data from the nitrate experiments in the region of low moderation.

^{3.} G. I. Bell, "A Method of Calculating Critical Masses of Proton Moderated Assemblies," LA-1548 (1953).

^{4.} C. K. Beck, A. D. Callihan, J. W. Morfitt, and R. L. Murray, "Critical Mass Studies, Part III," K-343 (1949).

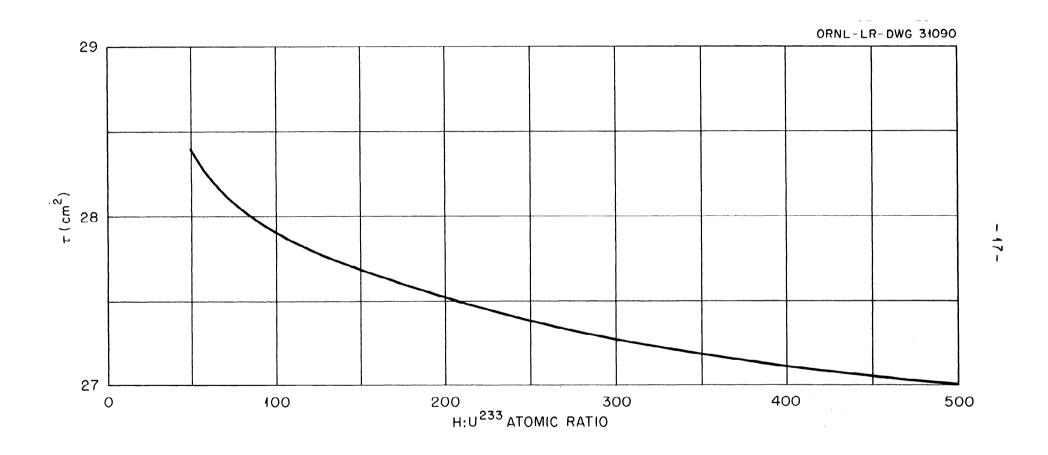


Fig. C-1. Variation of Neutron Age (τ) with the H:U 233 Atomic Ratio in Uranyl Nitrate Solutions

Since only two experimental determinations were made with unreflected vessels, one cylinder and one sphere, the extrapolation length adjusted to fit these points is highly uncertain. Similar comparisions using data for aqueous solutions of U^{255} in spheres indicate that the extrapolation length is always higher than for cylinders unless one applies a factor (0.965) to π^2 in the sphere buckling equation. If this factor is used, the extrapolation distance from the unreflected U^{255} sphere experiment agrees with that found with the cylinder. Figure C-2 is a plot of the calculated values of critical masses and volumes of unreflected cylinders using 4.95 cm for the extrapolation distance. These curves are only very rough approximations of these parameters since the supporting data are very limited.

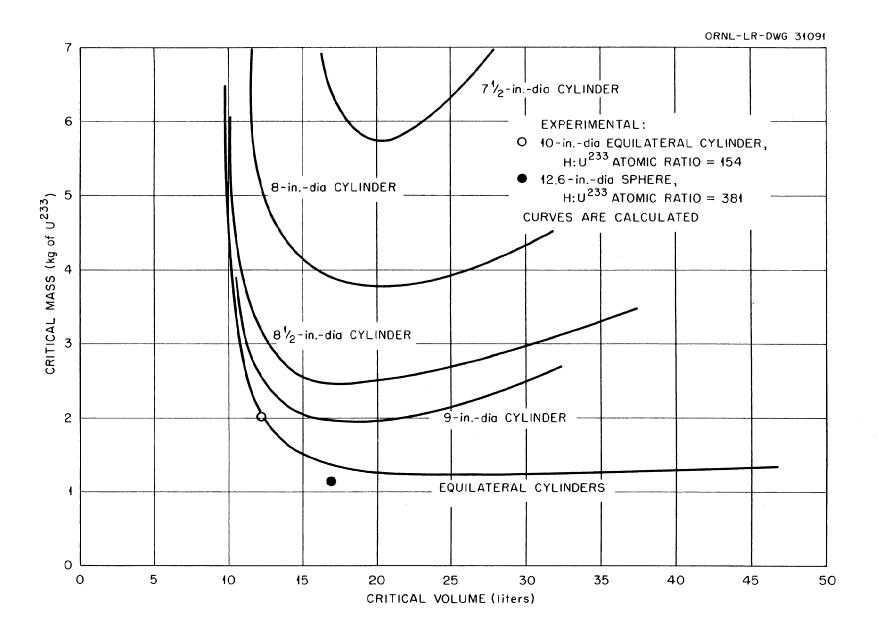


Fig. C-2. Estimated Critical Parameters of Unreflected Vessels Containing Aqueous Solutions of U 233

Appendix D

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